EFFICIENCY OF NEUTRON DETECTION OF SUPERHEATED DROPS OF FREON-22

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Abstract

Neutron detection efficiency of superheated drops of Freon-22 for neutrons obtained from a 3 curie Am-Be neutron source has been reported in this paper. Although Freon-22 having lower boiling point than many other similar liquids (e.g.Freon-12, Freon-114, Isobutane) is expected to be more sensitive to neutrons ,it has not been reported so far and therefore this paper constitutes the first report on the subject. Neutron detection efficiency of both Freon-22 and Freon-12 have been determined from the measured nucleation rate using the volumetric method developed in our laboatory. The result shows that neutron detection efficiency of Freon-22 for the neutron energy spectrum obtained from an Am-Be source, is almost double, while the life time is 58.6% smaller than that of Freon-12, for a particular neutron flux of that source.

1. INTRODUCTION

A liquid maintained at the same state above its boiling point is said to be superheated. It is a metastable state of the liquid and can be nucleated to form vapour by the deposition of small energy by ions, charged particles or by any heterogeneous nucleation sites such as gas pockets, impurities etc. The superheated drops,suspended in gel can be used to detect neutrons through the nucleation induced by the recoil nuclei in the medium . The recoil nuclei are produced by collision of neutrons with the nuclei constituting the superheated drops. The application of superheated drop detector (SDD) in neutron dosimetry has already been established (Apfel *et al.*, 1984, 1989; Ing, 1986) and several other potential applications of SDD in neutron research has been discussed (Apfel, 1979a, 1979b, 1981; Chakraborty *et al.*, 1990). Apfel (1992) has developed and characterised a passive superheated drop dosemeter using a volumetric technique for neutron monitoring of personnel and in accelerartor applications. These type of dosimeters are also commercially available from Apfel Enterprises Inc.,USA. Practical application of the SDD demands that the

sample must be reasonably stable against spontaneous nucleation due to background radiation and other environmental effects, and at the same time as much as possible sensitive to neutrons with energy spectrum of interest. Among the different important features of the detector, the systematic quantitative evaluation of the sensitivity of SDD has been studied for some liquids (e.g. Freon-114, Freon-12, Isobutane, Freon 142B) (Roy et al., 1987) and the response function was reported (Lo *et al.*, 1988). This paper is an attempt to investigate more sensitive liquid for neutron detection. Of the liquids investigated so far, Freon-12 is considered to be the most sensitive liquid for neutron detection. Since the boiling point of Freon-22 is much lower than Freon-12, we expect Freon-22 to be more sensitive to neutrons than Freon-12. To the best knowledge of the authors, no such investigation has been reported with Freon-22. An accurate method of determining the nucleation rate of SDD has been developed by Roy, *et al.*, (1997b) using a relative manometer. Some other studies on the neutron detection sensitivity and detector response were made by Ing and Birnboim (1984), Ing (1986), Ipe *et al.*, (1988) and Biro *et al.*, (1990). Nath *et al.*,(1993) measured the neutron dose equivalent to patients undergoing high energy x-ray and electron radiotherapy beams using a SDD device. They employed a passive method for measuring the total volume of neutron induced bubbles by displacing an eqivalent volume of gel into a graduated pipette. The method for the determination of efficiency of detection of neutrons by vapour nucleation of superheated drops using volumetric method has been developed in our laboratory by Roy *et al.*, (1997a). We in this work, present the efficiency of neutron detection, maximum nucleation rate and life-time of SDD of Freon-22 irradiated by neutrons from an Am-Be source for a particular neutron flux and compare it with those of Freon-12. Some of the physical parameters of SDD of Freon-12 and Freon-22 are presented in table-1. Comparison is made with Freon-12, since Freon-12 is the most well studied liquid in neutron detection. The paper has been organised to present a brief outline of the method of bubble formation, description of volumetric method and measurement, results and discussion.

2. THEORY OF BUBBLE FORMATION

The free energy required to form a spherical vapour bubble of radius r in a liquid is given by (Roy et al., 1987)

$$G = 4\pi r^2 \gamma(T) - \frac{4}{3}\pi r^3 (p_v - p_o) \tag{1}$$

where $\gamma(T)$ is the liquid-vapour interfacial tension,P_v is vapour pressure of the superheated liquid and P_o is the ambient pressure. The difference p_v -p_o is called the *degree of superheat* of a given liquid. One can see from equation (1) that G is maximum at

$$r = 2\gamma(T)/(p_v - p_o) = r_c \tag{2}$$

where r_c is called the *critical radius*. When a bubble grows to the size of the critical radius it becomes thermodynamically unstable and grows very fast till the entire liquid droplet vaporises. The minimum amount of energy (W) needed to form a vapour bubble of critical size r_c as given by Gibbs (1875) from reversible thermodynamics is

$$W = 16\pi\gamma^3(T)/3(p_v - p_o)^2 \tag{3}$$

which is supplied by the energy deposition dE/dx being the energy deposited per unit distance travelled by the nuclei in the liquid by the recoil nucleus in a path length of $2r_c$ inside the droplet.

3. PRINCIPLE OF THE VOLUMETRIC METHOD

The present method utilizes the superheated drops suspended in a dust free viscous elastic gel. The excess pressure required to form a vapour bubble of diameter 1mm inside this gel matrix is usually less than 1mm of mercury (as observed in a separate experiment). Hence the volume of the bubble trapped inside the gel and that at atmospheric pressure are almost equal. This is why the volume change upon nucleation would be the same whether the bubble formed is trapped inside the gel or liberated from it. Upon nucleation the increase in volume of the droplet would displace the trapped air inside a vial containing sample. In the present volumetric method described in detail in the next section, the measurement of rate of change of volume has been made using such an air displacement system. The superheated drops suspended in the gel do not touch each other physically and they nucleate in a random manner, independent of each other. Under such conditions it can be shown that the number of the drops and hence the volume of superheated liquid would decay

exponentially with time. The rate of change of nucleated volume varies exponentially with a time constant τ (lifetime). The life time of these droplets in presence of a neutron flux and the efficiency of neutron detection has been studied by measuring the volume of vapour formed upon nucleation.

If neutrons of flux ψ are incident on superheated drops of volume V, liquid density ρ_L and molecular weight M the vaporization rate is given by

$$\frac{dV}{dt} = V\psi\frac{N_A\rho_L}{M}\eta d\sum n_i\sigma_i \tag{4}$$

where N_A = Avogadro Number
d = average droplet volume
n_i = number of nuclei of the ith element of the molecule whose neutron nucleus elastic scattering cross section is σ_i
η = efficiency of neutron detection.

Due to nucleation by neutrons, the change in position (h) of the water column along a horizontal glass tube which is connected to the sample vial (arrangement is given in next section), could be measured with respect to time (t). The present set up is made completely free from any leakage. So the rate of increase of the volume of vapour during nucleation should be equivalent to the rate of decrease of the volume of the superheated liquid. So, we have this equation

$$\rho_V A\frac{dh}{dt} = -\rho_L\frac{dV}{dt} = -\rho_L V\psi\frac{N_A\rho_L}{M}\eta d\sum n_i\sigma_i \tag{5}$$

where A = cross section of the horizontal tube
ρ_V= density of Freon vapour
V = volume of superheated liquid at any instant of time t.

Integrating and solving equations (3) one can obtain

$$\frac{hA}{m} = a\left[1 - \exp -b\left(t - t_o\right)\right] \tag{6}$$

where a = $\frac{\rho_L V_o}{\rho_V m}$ and

$$b = \frac{1}{\tau} = \psi\frac{N_A\rho_L}{M}\eta d\sum n_i\sigma_i \tag{7}$$

$\frac{hA}{m}$ is the volume of accumulated vapour in time t per unit mass of the sample containing Superheated drops and gel, m being the mass of total sample (gel + superheated drops). Therefore the efficiency of neutron detection η is given by

$$\eta = \frac{bM}{\psi N_A\rho_L d\sum n_i\sigma_i} \tag{8}$$

V_o = initial volume of the Freon drops. t_o = initial time at which the experiment has been started. τ = *life-time* of SDD in presence of source. and the product ab gives the maximum nucleation rate. The equation (6) has been scaled to per unit mass of the sample for standardization in making comparison of two samples. Fitting equation (6) for different values of $\frac{hA}{m}$ and t, constants a, b are obtained. Knowing b, the life time τ (=1/b) in presence of neutron flux ψ is obtained. The neutron detection efficiency η can be found out from equation (8) for a known flux of neutrons

4. EXPERIMENT

The experiment was performed with 3Ci Am-Be neutron source, which has an energy distribution of neutrons with peak near 3.5 MeV. The source was placed inside a chamber through which neutrons coming in a fixed range of direction were used to irradiate the SDDs. Two separate sets of experiments were done with Freon-12 and Freon-22 respectively at about 30^oC and at atmospheric pressure. The superheated drops are usually suspended in an immiscible gel. The gel that we used here, was a homogeneous mixture of some ultrasonic gel and glycerol in suitable proportion. The detail of the preparation of the sample was given elsewhere (Roy et al., 1997b). The experimental apparatus consists of long glass tube of cross section 0.1573 sq.cm, placed horizontally on a graduated platform. The tube contained a coloured water column as an indicator of the volume of the vapour formed on nucleation. One end of the glass tube was connected to the glass vial containing the sample, by means of rubber tube. The glass vial and the horizontal tube were at the same height. So the pressure inside and outside the tube were equal, e.g. both were at the same atmospheric pressure. Therefore the displacement of the water column of 1 cm length due to nucleation would be directly related to the volume of the vapour formed. These displacements of the water column along the glass tube were measured as a function of time. The gamma sensitivity of sample of Freon-22 was tested with ^{241}Am (59.54keV), ^{60}Co (1170keV,1332kev), ^{137}Cs (662keV) gamma sources. The sample was placed either just in contact or at a close distance to the source. The sensitivity of the sample for 4.43MeV gamma ray present in ^{241}Am-Be source has also been tested by substantially reducing the neutron dose.

5. RESULTS

The chemical formula and other physical parameters including critical radius (r_c) and minimum energy required for nucleation (W) as calculated using equations

(2) and (3) respectively, for Freon-12 and Freon-22 have been listed in Table-1. The experimentally observed variation of volume of vapour formed, $\frac{hA}{m}$ (per unit mass of the sample) during nucleation as a function of time for Freon-12 and Freon-22 are shown in Fig.1 and Fig.2 respectively. The measured data on rate of nucleation and life-time of superheated drops of Freon-12 and Freon-22 in presence of neutrons from a 3 Ci Am-Be source and the efficiency of neutron detection are given in table-2. The samples irradiated by gamma sources did not show any noticeble nucleation at the experimental temperature.

6. DISCUSSION

From the tabulated results it is seen that the degree of superheat (defined by the difference of vapour pressure of the superheated liquid p_v and the ambient pressure p_o) attained for Freon-22 at room temperature (30^oC) and at atmospheric pressure is about 38% larger than that of Freon-12. This indicates that in presence of neutrons the life time of Freon-22 should be smaller than that of Freon-12. Our experimental results show that the maximum nucleation rate of Freon-22 is about 38% larger and the life time of Freon-22 is about 58.6% smaller than that of Freon-12 for a fixed neutron flux from an Am-Be source. As can be seen from the chemical formula that both of Freon-12 and Freon-22 contain carbon, chlorine and fluorine while Freon-22 contains one hydrogen replacing one chlorine in Freon-12. The neutron-nucleus elastic scattering cross-section for Freon-12 is 1.89 barns while that of Freon-22 is 1.69 barns at neutron energy 3.5 MeV. Although the probability of interaction of a neutron with nuclei of Freon-12 is larger than Freon-22, our experimental results (table-2) show that the present prepared sample of Freon-22 is about twice as efficient to detect neutrons than Freon-12. This is due to the fact that as seen from equation (3), the energy required for nucleation (W) decreases with degree of superheat and as is evident fron table-1, the saturation vapour pressure of Freon-22 at 30^oC is higher than that of Freon-12, therefore at this temperature Freon-22 will attain a higher degree of superheat which means a smaller amount of energy deposition is required for nucleation than that in Freon-12. So, although less number of recoil nuclei are available in Freon-22 from neutron nuclei elastic scattering, the percentage of nuclei capable of deposition of energy greater than W for Freon-22 must be larger than that in Freon-12. As a result the efficiency of neutron detection of SDD of Freon-22 is larger than that of Freon-12. From the experiment with gamma sources it is clear that the SDD based on Freon-22 is insensitive to gamma at the experimental temperature. The high efficiency of neutron detection and the insensitivity towards gamma makes Freon-22 a suitable superheated drop detector for neutron detection.

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Table-1 : A comparison of the physical parameters of Freon-12 and Freon-22

	Freon-12	**Freon-22**
1. ChemicalFormula	CCl_2F_2	$CHClF_2$
2. Molecular weight	120.91	80.47
3. Boiling Point	-29.79 ^{o}C	-40.75 ^{o}C
4. Surface tension (γ) (at 30^{o}C) dyn/cm	9	8
5. Vapour Pressure (p_v) dyn / sq. cm.	7.4556×10^6	1.14777×10^7
6. Density (ρ_L) gm / cc	1.293	1.175
7. Degree of superheat (p_v - p_o) dyn / sq. cm	6.441639×10^6	1.0463739×10^7
8. Critical radius $r_c = \frac{2\gamma(T)}{(p_v - p_o)}$ cm	2.79×10^{-6}	1.53×10^{-6}
9. Minimum energy required (W) keV to form a vapour bubble of size r_c ($W = \frac{16\pi\gamma^3(T)}{3(p_v-p_o)^2}$)	0.184	0.049

Table - 2: Observed results on nucleation
(Neutron flux = 2.5374×10^{7} /cm^2/s; Peak neutron energy = 3.5 MeV).

	Freon-12	**Freon-22**
1.Initial Nucleation rate[1] (cm^3/gm/s)	3.0268×10^{-4}	4.9205×10^{-4}
2. Lifetime τ (s)	3089.59	1279.13
3. Neutron detection efficiency[2]	0.2825%	0.5588%

1 Initial nucleation rate is $\left(\frac{1}{m}\right)\frac{dV}{dt}$ at the initial time, where m is the mass of the sample and $\frac{dV}{dt}$ is the rate of volume change upon nucleation.

2 neutron detection efficiency is the percentage of neutrons causing nucleation.

FIGURE CAPTIONS

Fig. 1: Observed variation of volume of vapour formed as a function of time for Freon-12; $\frac{hA}{m}$ = vapour volume per unit mass of the sample.

Fig. 2: Observed variation of volume of vapour formed as a function of time for Freon-22; $\frac{hA}{m}$ = vapour volume per unit mass of the sample.